ADDIS ABABA UNIVERSITY
SCHOOL OF GRADUATE STUDIES
ADDIS ABABA INSITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL AND BIO-ENGINEERING

Upgrading Biogas Produced from Biogas Pilot Plant through Absorption

A thesis Submitted to the Research and Graduate School of Addis Ababa University, Addis Ababa Institute of Technology, School of Chemical and Bio-Engineering in partial fulfillment of the requirements for the attainment of the Degree of Masters of Science in Chemical Engineering under Environmental Engineering Stream.

By: Gizachew Assefa Zeleke

Advisor: Dr.Ing Berhanu Assefa

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ADDIS ABABA, ETHIOPIA

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Approved by the Examining Board:

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Chairman, Department's Graduate Committee

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Advisor

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Internal Examiner

________________________ ________________

External Examiner

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List of abbreviations

AAiT	Addis Ababa Institute of Technology
AD	Anaerobic Digestion
ANOVA	Analysis of Variance
CD	Cow Dung
CDM	Clean Development Mechanism
CHP	Combined heat and power
CNG	Compressed Natural Gas
DM	Dry Mater
EC	European Calendar
EU	European Union
GF	Gas Flow rate
GHG	Greenhouse Gases
IADP	IGNIS Anaerobic Digestion Plant
LPG	Liquefied Petroleum Gas
MSW	Municipal Solid Waste
RHI	Renewable heat incentive
ROC	Renewable Obligation Certificate
RTFO	Renewable transport fuel obligation
UK	United Kingdom
VOC	Volatile Organic Carbon
WF	Water Flow rate
WH	Water Head

Abstract

Biofuels derived from biomass are considered as good alternative to petroleum fuels. Biogas is produced by anaerobic digestion of various organic substances such as food waste, agricultural waste, municipal solid waste (MSW), cow dung (CD) etc., which offers low cost and low emissions than any other secondary fuels. It can be a supplemented to liquefied petroleum gas (LPG) and compressed natural gas (CNG), if it is used in compressed form in cylinders. Ethiopia is among the developing countries where biogas upgrading system doesn't include in biogas production plants. The use of unrefined biogas has several social and environmental impacts such as GHG emissions, the incombustible part of biogas, CO_2, lowers its calorific value, the presence of hydrogen sulphide forms sulphuric acid with water, which is highly corrosive, rendering the biogas unusable. Furthermore, the use of biogas has remained near to the generating plants and compression requirement is high. This paper examines the effect of raw biogas flow rate, water flow rate and water head on methane enrichment of biogas; using water scrubbing which is based on the physical effect of dissolving gases in liquids. In a scrubber, CO_2 as well as the H_2S, dissolves into the water while CH_4 does not because of their difference in solubility. This makes it a very simple process. There is a lot of potentials in terms of increased energy per unit volume, wider application and ease of handling if biogas is upgraded to >95% CH_4. Raw biogas methane content in the studied pilot biogas plant ranged from 45 % to 56 %, carbon dioxide from 39 % to 50 % and trace amount of hydrogen sulphide. Oxygen content in all the measured raw biogas was < 2%. A counter-current absorption process upgraded the pilot plant gas near to 80 % methane content. The carbon dioxide content of the product gas ranged from 17 to 35 %. Hydrogen sulphide was removed from the raw pilot plant gas with over 90 % efficiency with the upgrading system. Removal of contaminants (CO_2 and H_2S) from biogas through upgrading was conducted at a water head of (80, 110 and 140 cm), water flow rate of (8, 16 and 25 l/min) and gas flow rate of (1, 1.5 and 2 l/min). In all cases, the increase in water head from 80 to 140 cm and the increase in water and gas flow rates increased the methane concentration. The highest methane content (80 %) was obtained at the highest water head (140 cm) and water flow (25 l/min) and when the gas flow was maintained at 1.5 l/min. The resulted CH_4 enriched product gas implies that carbon dioxide and hydrogen sulphide gases have been trapped in the absorbent water with higher removal efficiency (60 and >90 % respectively).

Keywords: biogas; methane; upgrading: Anaerobic digestion: enrichment: biofuel

1. INTRODUCTION

1.1 Background

Although world energy demand has continued to increase, supplies have dwindled. The present energy systems based on fossil fuel in addition to being finite, damage the natural environment in many and diverse ways, jeopardize human health and exert massive influences upon biogeochemical cycles. These have led credence to the need to turn energy systems towards sustainability.

There is large volume of organic solid waste generated and disposed into the field or landfill from time to time in urban. The organic waste produces a gas which is composed primarily of methane and carbon dioxide, greenhouse gases contributing to climate change. However, these wastes can be sorted and used for biogas plant feed. There are two major benefits of doing so. The first is that the biogas can be easily captured and prevents substances like methane from escaping to the atmosphere; and the second is that using biogas as a source of fuel and can replace the use of non-renewable sources of energy such as kerosene and natural gas.

Biogas was first used in Bombay in 1859 (IEA, 1997). Although this technology is known to the Chinese in 1950's and over hundred years old in India (Dioha *et al*, 2003), it is relatively recent in some developing countries. Biogas technology was introduced in Ethiopia as early as 1979, when the first batch type digester was constructed at the Ambo Agricultural College. In the last two and half decades around 1000 biogas plants, ranging in size from 2.5 m^3 to 200 m^3, were constructed in households, community and governmental institutions in various parts of the country. Presently, approximately 40% of the biogas plants that were constructed are not operational due to a lack of effective management and follow-up, technical problems, loss of interest, reduced animal holdings, evacuation of ownership, water problems, etc. (Eshete et al. 2008).

At the moment, biogas plants in developing countries do not include upgrading facilities. The implication is that all the CO_2, H_2S, other contaminants and their combustion products such as CO, SO_2, NOx, etc are released to the environment. With the sustained derive towards sustainability in energy systems in the country, the amount of these combustion products will increase and cannot be neglected.

Removing CO_2 and H_2S from the biogas is not easy. However, the upgrading technology is rapidly evolving, bringing biogas as a reliable energy source in sight (Saija, 2009). To produce large amounts of upgraded biogas, there are different upgrading methods (Hullu et al. 2008).In this study; we will look at the effectiveness of absorption using water as absorbent for improving the biogas energy content.

1.2 Problem Statement

Biogas which has a direct benefit for households in developing countries like Ethiopia, such as a low-cost fuel for cooking, reduced use of fuel food, improved soil fertility through the use of bio-slurry, job creations and as a source of fuel in gas engines to produce electricity in rural areas. Therefore, it is very important to develop inexpensive and efficient solution that enhances the purification of biogas by removing the large volume of CO_2 which reduces the heating value.

However, Ethiopia is among the developing countries where biogas upgrading system doesn't include in biogas production plants. The use of unrefined biogas has several social and environmental impacts such as GHG emissions, the incombustible part of biogas, CO_2, lowers its calorific value, the presence of hydrogen sulphide forms sulphuric acid with water, which is highly corrosive, rendering the biogas unusable.

Furthermore, the use of biogas has remained near to the generating plants and compression requirement is high. Special pipes would be required if raw biogas is to be distributed through gas pipelines due to the corrosive nature of H_2S content which also impacts unpleasant odour to the raw biogas (Glub and Diaz, 1991).

Many studies have proposed water scrubbing system as cheap and simple technology and also upgrading system may be incorporated in a family size plants. However, no scientific studies have been carried out on Ethiopian biogas upgrading until now. The motivation behind this research is to assess the absorption using water as a scrubbing solvent on pilot plant biogas production to see how enriched the biogas can be.

1.3 Objective of the Study

1.3.1 General Objective

To employ water scrubbing technology to scrub CO_2 and H_2S, so that biogas can be upgraded to a better biogas potential in terms of increased energy per unit volume.

1.3.2 Specific Objectives

- ☑ To identify parameters affecting biogas composition
- ☑ To Investigate the effects of water scrubbing on biogas composition
- ☑ To investigate the effect of upgrading on the energy content at IGNIS Biogas Pilot Plant
- ☑ To investigate the CO_2 and H_2S removal efficiency of the scrubbing process
- ☑ To evaluate efficiency of upgraded biogas

1.4 Significance of the Study

The significance of this study can be seen from different angles. In Ethiopia with a population of over 80 million and more than 90% of the energy demand of the country provided by biomass, a national biogas program is developed to promote the uptake of domestic biogas and to disseminate domestic biogas development as commercially viable market. Therefore, in order to efficiently use biogas, the nationwide biogas production facilities should be considered in parallel with the upgrading technologies. This research will have a role by putting one step forward the knowledge available around biogas upgrading.

2. LITERATURE REVIEW

2.1 Energy in Ethiopia

Access to modern energy is a key element in rural development. However, despite all attention given to energy issues in Ethiopia in the past, rural communities continue to be deprived of basic energy services. Modern forms of energy are simply not available in rural areas while traditional sources are rapidly being depleted, thereby deepening the rural energy crisis.

Among the principle manifestation of the rural energy crisis is depletion of wood fuel resources. This leads to the decline in household welfare caused by an increased use of inferior fuels, higher wood fuel prices, and a reduction in quality and frequency of cooked meals. Additionally, it leads to a reduction in agricultural productivity as a result of using dung and crop residue as fuel instead of using these as soil nutrients. Due to the use of dung as a source of domestic energy it is estimated that 10% of the annual grain production is lost for the Tigray region (Birhane *et al.*, 2005).

Ethiopia is endowed with various energy resources. The gross hydro-energy potential of the country is about 650 TWh per year, of which 25% could be exploited for power production (CESEN, 1986). This enormous potential ranks Ethiopia as one of the world's leading countries in hydro potential. The most promising hydropower development potential is found in the Blue Nile, Omo, and the Wabi Shebelle river basins (MEDaC, 1999). The energy potential of the country so far discovered comprises between 30 and 50 billion m^3 natural gas, more than 1000 MW geothermal power, and several hundred million tonnes coal and oil shale (Mariam, 1992). The total solar radiation reaching the territory is 2.3 TWh per year while wind energy potential is estimated at 4.8 million Tcal per year (CESEN, 1986). The country's woody biomass energy resources are about 14 million Tcal in standing stock and 0.93 million Tcal in terms of annual yield. The annual agricultural waste available for energy is about 176,000 Tcal per year.

Although the country has abundant energy resources, its potential is not yet well developed due to lack of capacity and investment. For example only less than 1% of the total hydropower potential of the country is known to have been utilized so far.

The energy sector in Ethiopia is composed of three main sub-sectors: biomass, petroleum and electricity. Energy consumption is very low, with an estimated total per capita consumption of only about 0.2 tonne oil-equivalent (Kidane et al. 2008).

2.2 Biogas

Biogas offers an attractive option to replace unsustainable utilization of wood and charcoal. Biogas is produced by methanogenic bacteria while acting upon biodegradable materials in an anaerobic condition. Biogas is mainly composed of 50 to 70% methane (CH_4), 30 to 40% carbon dioxide (CO_2) and small amounts of other gases (Lam and Heegde, 2007).

Biogas is generated in a biogas plant. The commonly used models are the floating drum, the fixed dome or Chinese model digester, the Deenbandhu model (also a fixed dome model) and the plastic bag digester.

The benefits of domestic biogas can be divided over multiple levels (micro, meso and macro) of the society and differ in the extent to which they can be translated in direct economic gains (formal versus informal) (Eshete *et al.*, 2006b) see Table 2.1.

Table 2.1: biogas benefit matrix (Kidane et al. 2008)

Informal		Formal
Micro	−Reduced indoor smoke-induced illnesses −Reduced poor-sanitation induced illnesses -Reduced drudgery from fuel wood collection -Reduced pressure for illegal forest encroachment -Reduced drudgery from weeding fields -Reduced workload for food-preparation -Reduced soil erosion/degradation -Improved opportunity for education -Reduced risk of erosion and landslides in mountainous areas	− Reduced direct medical costs − Reduced expenses on conventional energy sources − Reduced chemical fertilizer expenditures −Increased opportunity for (small-scale) organic agriculture −Improved agricultural yields −Increased family income −Increased employment and income-generating Opportunities
Meso	−Improved forest quality and quantity − Reduced pollution of the environment as a result of uncontrolled dumping of animal waste	− Opportunity to develop markets for (organic) agricultural products
Macro	− Reduction of illness-induced production losses −Improved biodiversity − Reduced mortality −Improved human resource base −Reduced risks resulting from global warming	− Reduced (forex) costs on medication − Reduced health system expenses − Reduced (forex) costs on chemical fertilizer − Reduced (forex) costs on fossil fuels − Increased agricultural production − Increased tax revenues − Generating CDM revenue

2.2.1 Composition of Biogas

2.2.1.1 Main Compounds of Biogas

Biogas from sewage digesters usually contains from 55 to 65 % methane, 35 to 45 % carbon dioxide and <1 % nitrogen, biogas from organic waste digesters usually contains from 60 to 70 % methane, 30 to 40 % carbon dioxide and < 1 % nitrogen, while in landfills methane content is usually from 45 to 55 %, carbon dioxide from 30 to 40 % and nitrogen from 5 to 15 % (Jonsson et al. 2003).

2.2.1.2 Trace Compounds of Biogas

Besides the main components, biogas also contains hydrogen sulphide and other sulphide compounds, siloxanes, aromatic and halogenated compounds. Although the amounts of trace compounds are low compared to methane, they can have environmental impacts such as stratospheric ozone depletion, the greenhouse effect and / or the reduction in local air quality. Also, some compounds cause engine failure if the gas is used as an energy source. Many volatile organic compounds (VOCs) with high vapour pressure and low solubility, which can occur in biogases, are harmful to the environment and / or to humans. Aromatics, heterocyclic compounds, ketones, aliphatics, terpenes, alcohols and halogenated aliphatics, for example, occur in particular in landfill gas (Allen et al. 1997, Eklund et al. 1998, Shin et al. 2002, Jaffrin et al. 2003). Many toxic VOCs are formed in household waste which includes cleaning compounds, pesticides, pharmaceuticals, plastics, synthetic textiles and coatings (Reinhart 1993).

2.2.1.2.1. Sulphur compounds

The main sulphur compound in biogases is hydrogen sulphide (H_2S). Other reduced sulphur compounds, as sulphides, disulphides and thiols, can also be found in biogases. In the presence of water, sulphur compounds can cause corrosion to compressors, gas storage tanks and engines (Persson et al. 2006), and thus these compounds need to be removed before biogas can be utilized as energy.

In biogas, in anaerobic conditions hydrogen sulphide and other sulphide compounds originate along several different pathways (Wilber & Murray 1990). For example, methanethiol and dimethyl sulphide (DMS) are formed from the degradation of sulphur-containing amino acids (e.g., in manure) and from the anaerobic methylation of sulphide. When DMS is reduced, following to methanogenic conversion, methane and methanethiol are formed. Methanethiol later forms methane, carbon dioxide and hydrogen sulphide (Lomans et al. 2002).

Properties of hydrogen sulphide

Physical and chemical properties

Hydrogen sulphide (H_2S) is a colourless, very poisonous gas. It is inflammable and forms explosive mixtures with air (oxygen). H_2S has a characteristic smell of "rotten eggs". This odour is only apparent in a small concentration range (0.05-500 ppm) (Navaratnasamy, 2008).

H_2S is soluble in water forming a weak acid. A combustion product of H_2S is SO_2. This makes the exhaust gases very corrosive (sulphuric acid) and contaminates the environment (acid rain).

Toxicity

H_2S is very poisonous (comparable to hydrogen cyanide). Lower toxic limit 10 ppm H_2S (Kapid *et a*l, 2004).

Effect

H_2S changes the red blood pigment; the blood turns brown to olive in colour. The transport of oxygen is hindered. The person suffocates "internally". The symptoms are irritation of the mucous membranes (including the eyes), nausea, vomiting, difficulty in breathing, cyanosis (discoloration of the skin), delirium and cramps, then respiratory paralysis and cardiac arrest. At higher concentrations immediate respiratory paralysis and cardiac arrest are the only symptoms. Even if a person survives poisoning, long term damage to the central nervous system and to the heart remains (Glub and Diaz, 1991).

The origins of hydrogen sulphide in biogas plants

Hydrogen sulphide is formed during microbiological reduction of sulphur containing compounds (sulphates, peptides, amino acids). Hydrogen sulphide is formed in the biogas plant by the transformation of sulphur-containing protein. This can be protein from plants and fodder residues. However, when animal and human feces are used, bacteria excreted in the intestines are the main source of protein. Inorganic sulphur, particularly sulphates, can also be biochemically converted to H_2S in the fermentation chamber (Helmut 1985).

Amounts

Plant material introduces little H_2S into biogas. On the other hand, poultry droppings introduce, on average, up to 0.5 vol. % H_2S, cattle and pig manure about 0.3 vol. % H_2S. Protein-rich waste (e.g. swill, molasses etc.) can produce large amounts of hydrogen sulphide (up to 3 vol. %). Inorganic sulphates (from salty, stall rinse water or diluting water) also produce considerable H_2S (Helmut 1985).

The effect of hydrogen sulphide on the biogas plant and the gas-utilization equipment

Fermentation inhibition

Dissolved H_2S is contained in the fermentation slurry. Equilibrium is set up between the dissolved H_2S and the H_2S in the gas phase. The dissolved H_2S in high concentrations can be toxic to the bacteria in the slurry. It can inhibit the production of biogas and cause its composition to alter (Helmut 1985).

Corrosion by H_2S

The presence of H_2S gas in biogas makes it corrosive to metal parts. Iron is subject to surface attack, although not major corrosion. Galvanized parts are similarly subject to surface corrosion. The effect on non-ferrous metals in components, such as pressure regulators, gas meters, valves and mountings, is much more serious. They are very quickly corroded. These materials also corrode in gas engines (seals and valves) (Kapid *et a*l, 2004).

Corrosion by SO_2 from H_2S

The combustion product SO_2 combines with water vapour and badly corrodes the exhaust side of burners, gas lamps and engines. Burning biogas in stoves and boilers can also result in damage to the chimney (Kapid *et a*l, 2004).

Engines

The acid which is formed corrodes engine parts in the combustion chamber, exhaust system and in various bearings. This is enhanced by frequent starts, short running times and the relatively low temperatures when starting up and after cutting off the engine. The water cooling system also provides the means (water needed to form sulphuric acid) for corrosion (Helmut 1985).

Engine oil changes

The suphur content of biogas used in gas engines shortens the time between oil changes. SO_2 from combustion and water vapour both dissolve in the lubricating oil. The oil becomes acidic and its properties change. It loses its ability to lubricate and sometimes corrodes metal components. Under continuous operating conditions the interval between oil changes is reduced to 200 - 250 hours (Helmut 1985).

2.2.1.2.2 Halogenated compounds

Halogenated compounds are often found in landfill gases and only rarely in biogases produced from sewage sludge or organic wastes. If biogas is used for energy production, compounds containing organochloride contribute to corrosion in vehicle or combustion engines while in certain combustion conditions the formation of dioxins and furans is also possible (Allen et al. 2007, Persson et al. 2006).

2.2.1.2.3 Organic silicon compounds

Organic silicon compounds present in biogas are oxidized during biogas combustion into microcrystalline silicon dioxide, as residue with chemical and physical properties similar to glass. Silicon dioxide collects in deposits on valves, cylinder walls, and liners, causing abrasion and blockage of pistons, cylinder heads, and valves. In gas turbines, siloxane deposits usually form on the nozzles and blades, causing erosion of the turbine blades and subsequently lowering operating efficiency (Tower 2003). Moreover, the glassy residues of silicon dioxide can de-activate the surface of the emission control system catalyst (Schweigkofler & Niessner 1999, Hagmann et al. 2002). Some organic silicon compounds can end up in the engine oil after the combustion process. In this case, the engine oil needs to be changed more frequently

(Environment Agency 2004). This has led some gas engine manufacturers to introduce a limit on silicon of 1 mg/ l in the oil of gas engines (Accettola & Haberbauer 2005).

2.3 Biogas Utilisation

Biogas can be used for all applications designed for natural gas. Different methods of environmental rating gave natural gas a 75% over all advantage over diesel and a 50% advantage over petrol (Nema and Bhuchner, 2002).

Upgrading raw biogas to >95% methane, positions the gas as an alternative vehicle fuel (Navaratnasamy, 2008), and reduces compression and storage requirements (Kapid *et a*l, 2004). The value added to biogas by upgrading and compression is as good as compressed natural gas; however upgraded biogas has the added advantage of lower emission profile associated with combustion (Nema and Bhuchner, 2002). Raw biogas is mainly used for heating and lighting. Combined heat and power (CHP) is used to convert biogas from fermentation tanks into electricity. However, in most cases, the heat is not efficiently used as a result of which about 60% of the energy is lost.

Table 2.2: Requirements to remove gaseous components depending on the biogas utilization (Kapid *et a*l, 2004)

Application	H_2S	CO_2	H_2O
Gas heater (boiler)	< 1000 ppm	No	No
Kitchen stove	Yes	No	No
Stationary engine (CHP)	< 1’000 ppm	No	no condensation
Vehicle fuel	Yes	recommended	Yes
Natural gas grid	Yes	Yes	Yes

2.3.1 Heating

Boilers do not have a high gas quality requirement. Gas pressure usually has to be around 8 to 25 mbar. It is recommended to reduce the H_2S concentrations to values lower than 1.000 ppm which allows to maintain the dew point around 150°C. The sulphurous acid formed in the condensate leads to heavy corrosion. It is therefore recommended to use stainless steel for the chimneys or condensation burners and high temperature resistant plastic chimneys. Most of the modern boilers have tin-laminated brass heat exchangers which corrode even faster than iron chimneys. Where possible, cast iron heat exchangers should be utilized. It is also advised to condense the water vapour in the raw gas. Water vapour can cause problems in the gas nozzles. Removal of water will also remove a large proportion of the H_2S, reducing the corrosion and stack gas dew point problems (Rautenbach *et al*, 2001).

2.3.2 CHP-engines

The utilization of biogas in internal combustion engines is a long established and extremely reliable technology. Thousands of engines are operated on sewage works, landfill sites and biogas installations. The engine sizes range from 45KW on small farms up to several MW on large scale landfill sites. Gas engines do have comparable requirements for gas quality as boilers except that the H_2S should be lower to guarantee a reasonable operation time of the engine. Otto engines designed to run on petrol are far more susceptible to hydrogen sulphide than the more robust diesel engines (Saija Rasi, 2009).

2.3.3 Vehicle fuel

The utilization of biogas as vehicle fuel uses the same engine and vehicle configuration as natural gas. In total there are more than 1 million natural gas vehicles all over the world, this demonstrates that the vehicle configuration is not a problem for use of biogas as vehicle fuel.

However, the gas quality demands are strict. With respect to these demands the raw biogas from a digester or a landfill has to be upgraded. Through upgrading we obtain a gas which has a higher calorific value in order to reach longer driving distances, has a regular/constant gas quality to obtain safe driving, does not enhance corrosion due to high levels of hydrogen sulphide, ammonia and water, does not contain mechanically damaging particles, does not give ice clogging due to a high water content, has a declared and assured quality. In practice this means that carbon dioxide, hydrogen sulphide, ammonia, particles and water (and sometimes other trace components) have to be removed so that the product gas for vehicle fuel use has methane content above 95 vol%.

In different countries different quality specifications for vehicle fuel use of biogas and natural gas are applied. Upgraded biogas is actually the cleanest vehicle fuel possible with respect to environment, climate and human health (Lntz et al. 2007).

2.3.3.1 Environmental benefits of bio-methane vehicles

Biogas produced from waste or energy crops can be upgraded and used as vehicle fuel in natural gas light-duty or heavy-duty vehicles. The same engine and vehicle configurations are used for biogas as for natural gas. In most cases, light-duty vehicles are so called “bi-fuel” vehicles, i.e. the vehicle can run either with petrol or gas. In some cases the vehicle is designed for natural gas or bio-methane and is optimized for operation on a single fuel. In heavy-duty vehicles, engines are usually designed to run solely on gas as the engines are based on the larger diesel engines, although “dual fuel” engines which use diesel as pilot ignition are being developed. In 2008 there were over nine million natural gas vehicles in the world, while the annual growth in the number of gas vehicles since the year 2000 has been 30%. In Asia, the annual growth has been over 50% and in Europe a little over 15% (Zittel 2008).

When biogas fuels are used as vehicle fuels typical air emission reductions compared to diesel fuel emissions have been from 60 to 85 % for nitrogen oxides, from 60 to 80 % for particulates and from 10 to 70 % for carbomonoxide (Schweigkofler & Niessner 2001).

If biogas were to replace petrol as a vehicle fuel, non-methane VOC emissions as well as the ozone forming potential of the engine emissions would decrease by 50 %. If biogas were to replace diesel as vehicle fuel, particulate emissions would be reduced significantly and in the case of heavy-duty vehicle, NOx emissions would decrease by 25 % (Jonsson et al. 2003).

2.4 Biogas Production Process

Bacteria decompose the organic matter in anaerobic environment. Biogas is an intermediate product of their metabolism. The decomposition process can be divided into 4 steps. Each of those are accompanied by different bacteria groups:

Hydrolysis

In the first stage aerobic bacteria reconstructs high molecular substances (protein, carbohydrates, fats, cellulose) by means of enzymes to low-molecular compounds like monosaccharide, amino acids, fatty acids and water (Yadvika et al., 2004). Enzymes assigned by hydrolysis bacteria decompose substrate components to small water-soluble molecules. Polymers turn into monomers (separate molecules). This process is called hydrolysis.

Acidogenesis

Then decomposition is made by acid-forming bacteria. Separate molecules penetrate into bacteria cells where further transformation takes place. This process is partially accompanied by anaerobic bacteria that consume the rest of oxygen hence providing suitable anaerobic environment for methane bacteria. The step is called acidogenesis as it is simply represented by the reactions below.

$C_6H_{12}O_6 + 2\ H_2O \rightarrow 2CH_3COOH + 2CO_2 + 4H_2$ (2.1)
(Acetic acid)

$C_6H_{12}O_6 \rightarrow CH_3CH_2CH_2COOH + 2CO_2 + 2H_2$ (2.2)
(butiric acid)

$C_6H_{12}O_6 + 2H_2 \rightarrow 2CH_3CH_2COOH + 2H_2O$ (2.3)
(propionic acid)

Acetogenesis

Afterwards acid-forming bacteria forms initial products for methane formation: acetic acid, carbon dioxide and hydrogen. These products are formed from organic acids. For vital functions of these bacteria that consume hydrogen, stable temperature mode is very important.

$CH_3CH_2COOH \rightarrow CH_3COOH + CO_2 + 3H_2$ (2.4)
(Acetic acid)

$CH_3CH_2CH_2COOH \rightarrow 2CH_3COOH + 2H_2$ (2.5)
(Acetic acid)

Methanogenesis

The last step is methane, carbon dioxide and water formation. Methane producing microorganisms occurs to the extent that anaerobic conditions are provided. 90% of methane yield takes place at this stage, 70% from acetic acid. Thus acetic acid formation (3rd step) is the factor that defines the speed of methane formations (ZORG Ukraine Biogas Plants, 2009, Hargono, 2007).

$CH_3COOH \rightarrow CH_4 + CO_2$ (2.6)
(Methane)

$2H_2 + CO_2 \rightarrow CH_4 + 2H_2O$ (2.7)

2.4.1 Factors Affecting Biogas Production

Nutrients

Organic matters, which are broken down by microorganisms without oxygen, will produce some quantities of methane. All biological process requires sufficient supply of nutrients particularly carbon and nitrogen as well as other elements are also required in trace quantities. The lack of specific elements required for microorganism growth will limit the production of biogas. Nutrients are assigned by the ratio of carbon and nitrogen (Anunputtikul, 2004).

PH

Hydrocarbons are easier to acidify, and no pH-buffering ions are released as with the degradation of proteins. Therefore the pH-value decreases more easily. With the degradation of carbohydrates, the partial pressure of hydrogen increases more easily, as with other substances. This happens in combination with the formation of reduced acidic intermediate products. The pH optimum of the methane-forming microorganism is at pH = 6.8-7.2. Therefore, it is important to adjust the pH-value. Only Methanosarcina is able to withstand lower pH values (pH = 6.5 and below). With the other bacteria, the metabolism is considerably suppressed at pH <6.7 (Dieter Deublein, 2008).

Temperature

Two optimum temperature levels have been established the mesophilic level (25-40^{0}C) and thermophilic level (50-65^{0}C) (Bitton, 1994; Busby *et al*., 1977; Mackie and Bryant. 1995). Most of the methanogenic microorganisms belong to the mesophilics. Only a few are thermophilic. Methanogenics are sensitive to rapid changes of temperature. Thermophilic methanogens are more temperature-sensitive than mesophilics. Even small variations in temperature cause a substantial decrease in activity. Therefore, the temperature should be kept exactly within a range of +/-2°C. Under mesophilic operating conditions, the inhibition of ammonium is reduced because of the lower content of inhibiting free ammonia. In general, it has to be mentioned that the energy balance is better in the mesophilic range than in the thermophilic range (Dieter Deublein, 2008.

2.5 Biogas Upgrading and Bio-Methane Production Technologies

In order to guarantee that a gas product can safely and efficiently be used in vehicles as a Bio-methane bio-fuel or to be injected into the natural gas network the bio-methane must meet a gas quality specification. Upgraded biogas (bio-methane) has similar properties to natural gas, which has been used as a vehicle fuel for decades. To utilize bio-methane as a fuel for vehicles, the same engine and vehicle configuration that is used for natural gas can be used.

In 1999 Sweden developed a national standard for the use of biogas as a vehicle fuel upon request of the Swedish motor industry (Persson et al., 2006). The parameters are summarized in Table 2.3.

Table 2.3: Swedish Standard for bio-methane (Persson et al., 2006)

Parameter	Requirement
Lower Wobbe Index	43.9 – 47.3 MJ/Nm^3
Methane concentration	97±2% by volume
Motor Octane Number	>130
Water dew point	t-5, where t = ambient temperature
$CO_2+O_2+N_2$	<5% by volume
O_2	<1% by volume
Total sulphur	<23 mg/Nm^3
NH_3	<20 mg/Nm^3

This standard is similar to that applied when injecting bio-methane into the natural gas network.

In order to use biogas as a vehicle fuel or for injection into the natural gas network the biogas must be cleaned (the CO_2 and H_2S removed) and upgraded (>95% methane) to produce bio-methane, which has a higher calorific value, to provide longer driving distances, or an energy content corresponding with the gas distributed via the natural gas grid. The quality standards ensure that the bio-methane does not aggravate corrosion, does not contain mechanically damaging particles, and has a declared and assured quality (eurObserv'ER, 2008).

Biogas upgrading processes need to be designed to avoid or minimize emissions. For example, in absorption processes some of the methane can be absorbed to the absorption liquid and be released into the air with the exhaust gas, hydrogen sulphide, other reduced sulphur compounds and halogenated compounds in exhaust gas can have environmental and health risks if released into the air untreated. The absorption liquid used in the process should also be treated, for example with other wastewaters (Environmental Agency, 2004)**.**

A number of gas upgrading technologies have been developed for the treatment of natural gas, town gas, sewage gas, landfill gas etc. However, not all of them are recommended for the application with biogas because of price and/or environmental concerns. It is clearly seen in (Table 2.4) that water scrubbing is preferably used because of its feasibility.

Table2.4: List of selected reference plants with full gas upgrading to natural gas / vehicle fuel standards

Country	City	Product gas utilization (gas grid or vehicle fuel)	Biogas production (landfill/sewag	CH_4 requirement %	CO_2 removal technique	H_2S removal techniqe	In operation since
Czech Rep.	Bys-tran	Vehicle fuel	Sewage sludg	95	Water scrub.	Water scrub.	1985
	Byst-rica	Vehicle fuel	Sewage sludge	95	Water scrub.	Water scrub.	1990
	Chanov	Vehicle fuel	Sewage sludge	95	Water scrub.	Water scrub.	1990
	Liberec	Vehicle fuel	Sewage sludge	95	Water scrub.	Water scrub.	1988
	Zlin/Teco-Vice	Vehicle fuel	Sewage sludge	95	Water scrub.	Water scrub.	1990
France	Cham-béry	Vehicle fuel	Sewage sludge	96.7	Water scrub.	Boil.filter/water scrub.	-
	Lille	Vehicle fuel	Sewage sludge	-	Water scrub.	Water scrub.	1995
	Tours	Vehicle fuel	Landfill	-	Water scrub.	Water scrub.	1994
	Collendorn	Natural gas	Landfill	88	Membranes	Activated carbon	1991
	Gorredijk	Natural gas	Landfill	88	Membranes	Activated carbon	1994
The Netherlands	Nuenen	Natural gas	Landfill	88	Carbon molecular sieves	Activated carbon	1990
	Tilburg	Natural gas	Sewage sludge/landfill/green waste	88	Water scrub.	Iron oxide pellets	1987
	Wijster	Natural gas	Landfill	88	Carbon molecular sieves	Activated carbon	1989
New Zeeland	Christ-Church	Vehicle fuel			Water scrub.	Water scrub.	
Sweden	Eslöv	Vehicle fuel	Sewage sludge/ vegetable waste	97	Water scrub.	Activated carbon	1998
	Göteborg	Vehicle fuel	Sewage sludge	97	Carbon molecular sieves	Water scrub.	1992
	Helsin-Gborg	Vehicle fuel	Slaughter house waste	97	Carbon molecular sieves	Water scrub.	1996
	Kalmar	Vehicle Fuel	Sewage sludge +manure	97	Water Scrub.	Iron chloride dosing+ water scrub.	1998
	Linköping	Vehicle fuel	Sewage sludge +manure	97	Water scrub.	Water scrub.	1997

2.5.1 Drivers behind the Development of Biogas Upgrading Technologies to Produce Biomethnae both as a Vehicle Fuel and for Injection into the Gas Grid

2.5.1.1 Political Drivers

EU policy targets require the use of 20% of renewable energy by 2020 and a cut in carbon dioxide emissions by 2050 of up to 60% (European Parliament 2008). Biogas can provide renewable energy in the form of electricity, heat, and transport fuel. The generation of renewable electricity from biogas is well established, but the enhancement of biogas to bio-methane for use as a vehicle fuel or for injection into the gas grid is increasingly being promoted (Hongo 2008).

Member states are obliged to ensure that bio-methane has the same access to the gas grid as natural gas, provided that it meets the quality and safety requirements and that a continuous supply can be guaranteed. However, only some countries have currently adopted safety standards for the injection of bio-methane into the gas grid. At the same time EU Directive 2003/30/EC obliges member states to develop and implement policies to promote the use of bio-fuel in the transport sector. The goal is to replace 5.75% of transport energy use with bio-fuel by 2010 (Hongo 2008).

2.5.1.2 Economic Drivers

There are two main economic drivers that may promote the utilization of biogas as renewable energy: government incentives and operational profit. To date the renewable energy market in Europe is clearly dominated by the use of CHP engines to produce heat and electricity. This is encouraged by government incentives, such as financial benefits and grant support (Lantz et al. 2007).

Most countries pay the renewable energy producer a commodity amount per kWh of electricity and/ or heat produced. For example in the UK, this is currently two times one “ROC” (Renewable Obligation Certificate), with each valued at 3.7p/kWh plus a small payment associated with the Climate Change Levy, which together totals 7.8p/kWhr (8.7ct/kWhr), for renewable electricity produced. In Germany, the equivalent incentive is 11.67ct/kWh. This payment is in addition to the electricity and heat sales price, thus allowing the operators to realize considerable revenue from energy production (Lantz et al. 2007).

Compared to the CHP sector, the conversion of biogas to bio-methane is still a novelty in many EU countries, and economic incentives are not widespread. Sweden is pioneering the use of biogas as vehicle fuel and has introduced a range of incentives, including tax benefits, free parking, and the exemption of congestion charges for bio-methane fuelled vehicles.

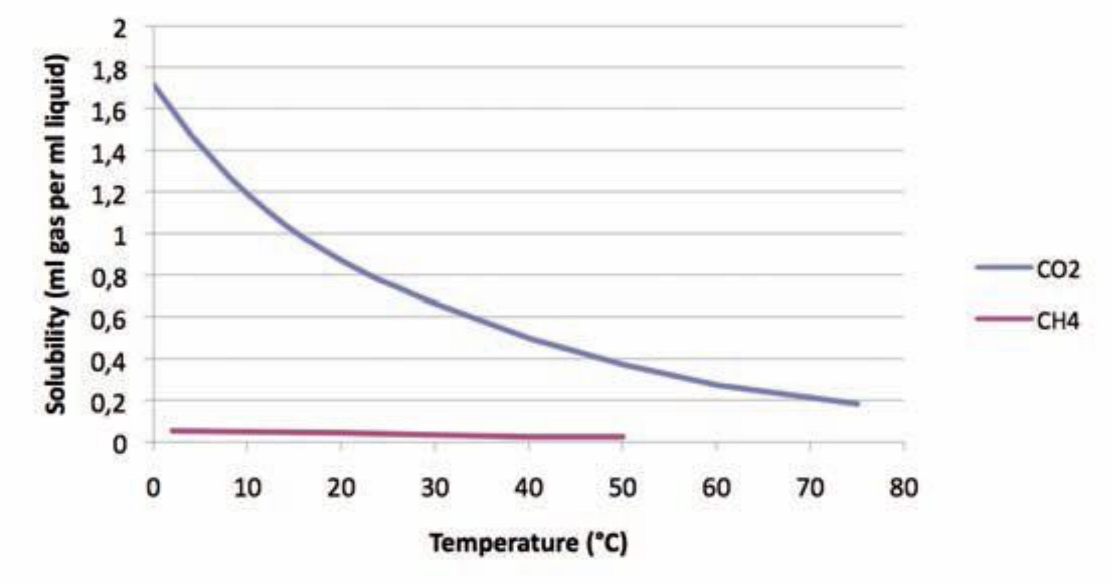
Additionally, it was proposed in 2007 to have at least one renewable fuel pump at every petrol station in the country. Other countries, particularly Germany and Switzerland, recently also started putting increased efforts into the development and use of bio-methane as a fuel, being encouraged by fuel incentives, tax exemptions and European legislation, but also by rising energy prices. The German government pays the operators a baseline bonus of 8ct/kWh for the utilization of energy from biomass, a technology bonus of 2ct/kWh for biogas upgrading (both as vehicle fuel or for injection into the gas grid) and also offers grants of 30% of the capital investment for a biogas upgrading plant. Additionally, biogas fuelled vehicles are tax exempt until at least 2015. Similar tax incentives are now being introduced by the UK government. From April 2011 there will be feed-in tariffs for heat under the renewable heat incentive (RHI), which will also apply to bio-methane injected into the gas grid. A similar incentive for vehicle fuel, the renewable transport fuel obligation (RTFO), will come into effect for biogas upgraded as vehicle fuel, although the rate of this incentive has not yet been set by the UK government.

2.5.2 Biogas Upgrading Technologies

2.5.2.1 Absorption

The separation principle of absorption is based on different solubilities of various gas components in a liquid scrubbing solution. In an upgrading plant using this technique the raw biogas is intensively contacted with a liquid within a scrubbing column filled with packing in order to increase the contact area between the phases.

Carbon dioxide has a higher solubility in water than methane. Carbon dioxide will therefore be dissolved to a higher extent than methane, particularly at lower temperatures (Figure 2.1). As a result, the remaining gas stream is enriched with methane and the scrubbing liquid leaving the column is rich in carbon dioxide. In order to maintain the absorption performance, this scrubbing liquid has to be replaced by fresh liquid or regenerated in a separated step (desorption or regeneration step).



Currently, three different upgrading technologies embodying this physical principle are available.

2.5.2.1.1 Physical absorption: Water scrubbing

Water scrubbing is used to remove carbon dioxide and also hydrogen sulphide from biogas since these gases are more soluble in water than methane. The absorption process is purely physical. Usually the biogas is pressurized and fed to the bottom of a packed column where water is fed on the top and so the absorption process is operated counter-currently.

Water scrubbing can also be used for selective removal of hydrogen sulphide since hydrogen sulphide is more soluble than carbon dioxide in water. The water which exits the column with absorbed carbon dioxide and/or hydrogen sulphide can be regenerated and recirculated back to the absorption column. The regeneration is made by de-pressurizing or by stripping with air in a similar column. Stripping with air is not recommended when high levels of hydrogen sulphide are handled since the water will soon be contaminated with elementary sulphur which causes operational problems. The most cost efficient method is not to recirculate the water if cheap water can be used, for example, outlet water from a sewage treatment plant.

The absorbed gas components are physically bound to the scrubbing liquid, in this case water. Carbon dioxide has a higher solubility in water than methane and will therefore be dissolved to a higher extent, particularly at lower temperatures and higher pressures. In addition to carbon dioxide, also hydrogen sulphide and ammonia can be reduced in the bio-methane stream using water as a scrubbing liquid.

The effluent water leaving the column is saturated with carbon dioxide and is transferred to a flash tank where the pressure is abruptly reduced and the major share of the dissolved gas is released. As this gas mainly contains carbon dioxide, but also a certain amount of methane (methane is also soluble in water, but to a smaller extent) this gas is piped to the raw biogas inlet.

The drawback of this method is that the air components oxygen and nitrogen are dissolved in the water during regeneration and thus, transported to the upgraded bio-methane gas stream. Therefore, bio-methane produced with this technology always contains oxygen and nitrogen. As the produced bio-methane stream is saturated with water, the final step in upgrading typically is gas drying, for example by the application of glycol scrubbing.

The rate of CO_2 and H_2S absorption depends upon the factors such as,

- Gas flow pressure
- Composition of biogas
- Water flow rates
- Purity of water and
- Dimension of scrubbing tower

There are two types of water scrubbing

Single pass scrubbing

In single pass scrubbing the washing water is used only once. The advantage of this type of scrubbing is that there is no contamination of the water with traces of H_2S and CO_2. This gives that the total amount of CO_2 and H_2S is at its maximum.

The disadvantage of this technique is that it requires a large amount of water. This technique is only feasible when working near a sewer water cleaning plant of which water can be used.

Regenerative absorption

In regenerative absorption, the washing water is regenerated after washing the biogas. The main advantage of this technique is that the total amount of water required is much lower compared to single pass scrubbing.

Water scrubbing requires a large amount of water. How much H_2S and CO_2 can be dissolved is rather important for the design of a water scrubber. The dissolubility of H_2S and CO_2 increases with increasing pressure. This relation can be described by Henry's Law:

$$Pi = H.\, C_{max} \qquad (1)$$

Where

C_{max} -Saturation concentration of the component [mol/m^3]

H - Henry's coefficient [$Pa.\, m^3/mol$]

Pi - Partial pressure of the component [Pa]

According to Dalton's law, the total pressure is the sum of all partial pressures. So, if the total pressure is increased, the partial pressure increases the same factor. This gives that the saturation concentration rises as well.

However, when higher pressures are reached the dissolubility of the components will not linearly increase with the pressure. At higher pressures, the increase of dissolubility becomes lower. Until a pressure of 20 bar the dissolubility can be described according to Henry's law (Alfons, 2006). These calculations are based on the ideal situation so non-idealities should be taken into account when designing the scrubber.

The mass transfer of components from the gas phase to the water phase and vice versa is important to know. When this is known, the dimensions of the reactor can be calculated. Mass transfer occurs when a high concentration difference between two phases is realized. The mass transfer can be described using the double film model.

When two layers with different concentration profiles intersect, the following equations can be written:

$$N_{AG} = k_G \,.\, a. (C_{AG} - C_{AGi}) \qquad (2)$$

$$N_{AL} = k_L \,.\, a. (C_{ALi} - C_{AL}) \qquad (3)$$

The mass transfer coefficients, k_L and k_G, are dependent on a lot of parameters. It is difficult to get a precise measurement of these values. But when there is a general idea of this value, the dimensions of the scrubber can be designed.

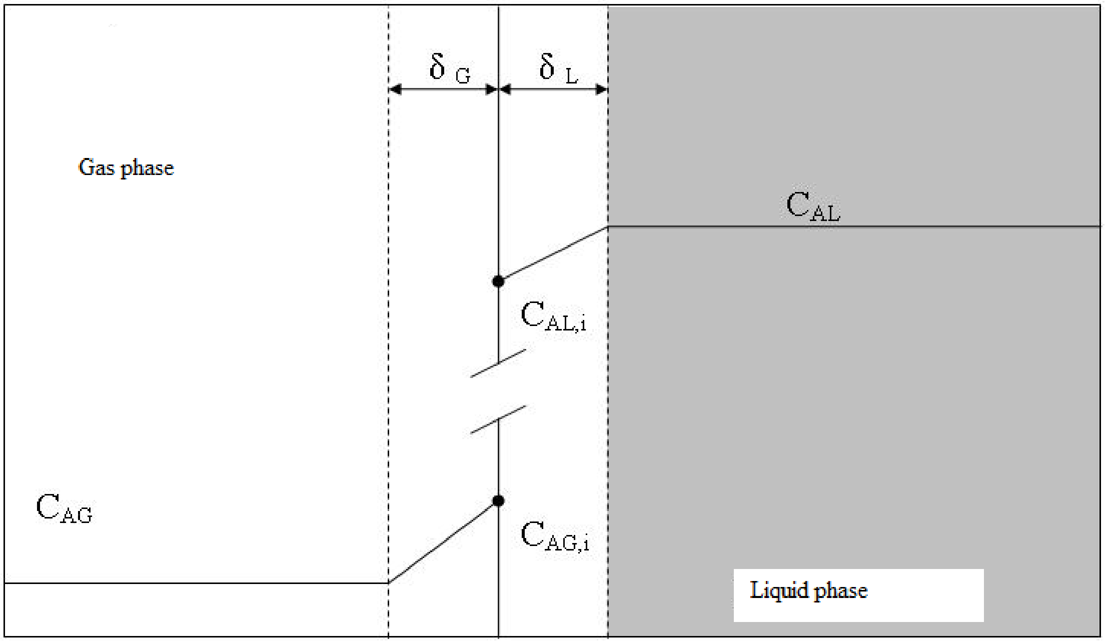


Figure 2.2 Concentration profile in double film model

Water scrubbing is the simplest way to upgrade biogas. It is this simple because it only requires water and an absorption column to upgrade the biogas.

2.5.2.1.2 Organic physical absorption

Very similar to water scrubbing, this technology uses an organic solvent solution (e.g. polyethylene glycol) instead of water as a scrubbing liquid. Carbon dioxide shows higher solubilities in these solvents than in water. As a result, less scrubbing liquid circulation and smaller apparatuses are needed for the same raw biogas capacity.

2.5.2.1.3 Chemical absorption

Chemical absorption is characterized by a physical absorption of the gaseous components in a scrubbing liquid followed by a chemical reaction between scrubbing liquid components and absorbed gas components within the liquid phase. As a result, the bonding of unwanted gas components to the scrubbing liquid is significantly stronger and the loading capacity of the scrubbing liquid is several times higher.

The chemical reaction is strongly selective and the amount of methane also absorbed in the liquid is very low resulting in very high methane recovery and very low methane slip. Due to the high affinity of especially carbon dioxide to the used solvents (mainly aqueous solutions of

Monoethanolamine MEA, Diethanolamine DEA and Methyldiethanolamine MDEA) the operating pressure of amine scrubbers can be kept significantly smaller compared to pressurized water scrubbing plants of similar capacity.

Chemical absorption involves the formation of reversible chemical bonds between the solute and the solvent. Regeneration of the solvent, therefore, involves breaking of these bonds and correspondingly, a relatively high energy input. Chemical solvents generally employ either aqueous solutions of amines, i.e. mono-, di- or tri-ethanolamine or aqueous solution of alkaline salts, i.e. sodium, potassium and calcium hydroxides.

2.5.2.1.4 Polyethylene glycol scrubbing

Polyethylene glycol scrubbing is like water scrubbing a physical absorption process. Selexol is one of the trade names used for a solvent. In this solvent, like in water, both carbon dioxide and hydrogen sulphide are more soluble than methane. The big difference between water and Selexol is that carbon dioxide and hydrogen sulphide are more soluble in Selexol which results in a lower solvent demand and reduced pumping. In addition, water and halogenated hydrocarbons (contaminants in biogas from landfills) are removed when scrubbing biogas with Selexol. Selexol scrubbing is always designed with recirculation. Due to formation of elementary sulphur stripping the Selexol solvent with air is not recommended but with steam or inert gas (upgraded biogas or natural gas). Removing hydrogen sulphide on beforehand is an alternative.

2.5.2.2 Adsorption

In adsorption processes, the solute (CO_2) in the raw biogas adsorbs into the surface of a solid material usually by means of weak Van der Waal Forces, London Forces etc. Almost all solids can adsorb, however granular solid with large surface area per unit volume are employed as commercial adsorbents. Good adsorbents are capable of removing CO_2, H_2S, moisture and other impurities from biogas. Solid adsorbents used for purification of gas include alumina activated carbon, silica, etc., and are referred to as molecular sieves. Adsorption systems are simple in design and easy to operate. Although they have good moisture holding capacity, the high heat and pressure required make them expensive process.

2.5.2.2.1 Pressure swing adsorption (PSA)

With this technique, carbon dioxide is separated from the biogas by adsorption on a surface under elevated pressure. The adsorbing material, usually activated carbon or zeolites, is regenerated by a sequential decrease in pressure before the column is reloaded again, hence the name of the technique. An upgrading plant, using this technique, has four, six or nine vessels working in parallel. When the adsorbing material in one vessel becomes saturated the raw gas flow is switched to another vessel in which the adsorbing material has been regenerated. During regeneration the pressure is decreased in several steps. The gas that is desorbed during the first and eventually the second pressure drop may be returned to the inlet of the raw gas, since it will contain some methane that was adsorbed together with carbon dioxide. The gas desorbed in the following pressure reduction step is either led to the next column or if it is almost entirely methane free it is released to the atmosphere. If hydrogen sulphide is present in the raw gas, it will be irreversibly adsorbed on the adsorbing material. In addition, water present in the raw gas can destroy the structure of the material. Therefore hydrogen sulphide and water needs to be removed before the PSA-column.

2.5.2.2.2 Adsorption on activated carbon

Hydrogen sulphide is adsorbed on the inner surfaces of engineered activated carbon with defined pore sizes. Addition of oxygen (in the presence of water) oxidizes H_2S to plane sulphur that binds to the surface. In order to increase the speed of the reaction and the total load, the activated carbon is either impregnated or doped (by addition of a reactive species before formation of the activated carbon) with permanganate or potassium iodide (KI), potassium carbonate (K_2CO_3) or zinc oxide (ZnO) as catalysers. For grid injection or utilisation as vehicle fuel, only marginal amounts of O_2 are allowed in the gas. Hence oxidation of the sulphur is not suitable. In those cases mostly KI-doped carbon or permanganate impregnated carbon is used because addition of oxygen is not required in the case of KI under reduced loading. While ZnO impregnated carbon is rather expensive, H_2S removal is extremely efficient with resulting concentrations of less than 1ppm.

2.5.2.3 Membrane purification technology

Membranes for biogas upgrading are made of materials that are permeable for carbon dioxide, water and ammonia. Hydrogen sulphide, oxygen and nitrogen permeate through the membrane to a certain extent and methane passes only to a very low extent. Typical membranes for biogas upgrading are made of polymeric materials like polysulfone, polyimide or polydimethylsiloxane. These materials show favorable selectivity for the methane/carbon dioxide separation combined with a reasonable robustness to trace components contained in typical raw biogases. To provide sufficient membrane surface area in compact plant dimensions these membranes are applied in form of hollow fibers combined to a number of parallel membrane modules (Natcogroup, 2007).

After the compression to the applied operating pressure the raw biogas is cooled down for drying and removal of ammonia. After reheating with compressor waste heat the remaining hydrogen sulphide is removed by means of adsorption on iron or zinc oxide. Finally, the gas is piped to a single- or multi-staged gaspermeation unit. The numbers and interconnection of the applied membrane stages are not determined by the desired bio-methane quality but by the requested methane recovery and specific compression energy demand. Modern upgrading plants with more complex design offer the possibility of very high methane recoveries and relatively low energy demand. Even multi-compressor arrangements have been realized and proved to be economically advantageous. The operation pressure and compressor speed are both controlled to provide the desired quality and quantity of the produced bio-methane stream.

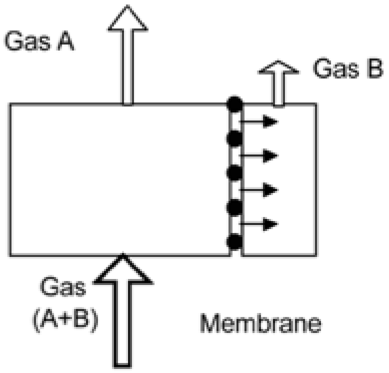


Figure 2.3: Schematic representation of membrane separation (M. Harasimowicz et al. 2007)

Any polymeric material will separate gases to some extent. Proper selection of the polymeric material comprising the membrane is extremely important. It determines the ultimate performance of the gas separation module. Membranes made of polymers and copolymers in the form of a flat film or a hollow fibre have been used for gas separation. Several different membranes have been found in literature. The Natcogroup use cellulose acetate as a base membrane material (Natcogroup, 2007). Cellulose acetate is very inert and stable in CO_2 hydrocarbon environments. Application of polyimide membranes has also been found (M. Harasimowicz et al. 2007). For this type of membrane, a single stage unit is sufficient to achieve 94% enrichment from gas with a common concentration of CH_4.

2.5.2.4 Biological processes

Hydrogen sulphide can be oxidized by microorganisms of the species Thiobacillus and Sulfolobus. The degradation requires oxygen and therefore a small amount of air (or pure oxygen if levels of nitrogen should be minimized) is added for biological desulphurization to take place. The degradation can occur inside the digester and can be facilitated by immobilizing the microorganisms occurring naturally in the digestate. An alternative is to use a trickling filter which the biogas passes through when leaving the digester. In the trickling filter the microorganisms grow on a packing material. Biogas with added air meets a counter flow of water containing nutrients. The sulphur containing solution is removed and replaced when the pH drops below a certain level. Both methods are widely applied, however they are not suitable when the biogas is used as vehicle fuel or for grid injection due to the remaining traces of oxygen.

2.6 Comparison of different upgrading techniques

The most widely used technologies for biogas upgrading are pressure swing adsorption, water scrubbing, organic physical scrubbing and chemical scrubbing. Their characteristics as given by the technology providers are summarized in Table2.5. However, it is important to remember that the best technology to choose is based on specific parameters at the plant, such as the availability of cheap heat and the electricity price. It should also be noted that it is often possible to lower the methane loss, but at the expense of a higher energy consumption.

Today, technological developments have led to cheaper and more efficient plants thanks to the increasing interest in upgrading biogas. The demand for more plants has also led to the development of standardized upgrading units which also decreases the costs.

The upgrading costs of established techniques are dependent on the specific technology, but most importantly on the size of the plant. However, the field of biogas upgrading is developing rapidly and thus the cost development would also be expected to change. Today, there are commercially available plants for capacities lower than 250 Nm3/h, while also plants larger than 2000 Nm3/h are being built. These developments and also the fact that more plants are being built will likely lead to lower prices.

Table2.5: Comparison between selected parameters for common upgrading processes (Urban et al. 2008)

Parameter	PSA	Water scrubbing	Organic physical scrubbing	Chemical scrubbing
Pre-cleaning needed	Yes	no	no	Yes
Working pressure (bar)	4-7	4-7	4-7	No pressure
Methane loss	< 3 % / 6 –10%	< 1 % / < 2%	2– 4%	< 0.1%
Methane content in upgraded gas	> 96%	> 97%	> 96 %	> 99%
Electricity consumption (kWh/Nm3)	0.25	< 0.25	0.24–0.33	< 0.15
Heat requirement (°C)	N0	No	55–80	160

3. MATERIALS AND METHODS

3.1 Materials

3.1.1 IGNIS Biogas Pilot Plant

In this research, the biogas produced by IGNIS Biogas Pilot Plant**,** an anaerobic digestion (AD) plant was used for the upgrading experiment. The AD plant had been set up to analyze the feasibility of the anaerobic digestion of biological waste in Addis Ababa, in particular food left over (Figure 3.1). The volume of the fermenter is 15m^3, with a daily DM input of 54 kg. The digester load is 3.6 Kg DM/ (m^3 d). It is expected that the daily input will turn into 28.7Nm^3 of biogas per day or 1.2Nm^3/h. The fermenter was designed for a daily input of 200 Kg of solid waste and 400 l of wastewater. Table 3.1 shows the expected amount of daily delivered substrate, the dry mater (DM) content of the substrate and the calculated amount of biogas.

The fermenter is insulated and kept at a temperature between 37^{O}C and 40^{O}C. The necessary heat is provided by two solar collectors (total collector surface = 4m^2) that are installed at the roof of a container.

The hot water from the collectors is stored in a buffer tank (800 l). The buffer tank ensures a full time heat supply. An electric heater is installed as an emergency heating and to provide the necessary heat during the rainy season.

To this effect, the plant had been erected in proximity to a student canteen, to use kitchen waste and kitchen wastewater as a feed for biogas pilot plant (Katharina Panse 2013). This research is planned to improve the quality of biogas produced from this pilot plant.

Table 3.1: Added amount of waste and expected daily biogas quantity (Katharina Panse 2013)

Substrate	Weight [kg/d]	DM [%]	DM [kg/d]	Biogas [Nm^3/d]	Digestate [t/a]
Wastewater	400	2	8	2.7	144.69
Solid Waste	200	23	46	26	59.44
Total	600	25	54	28.7	204.13

Figure 3.1: IGNIS anaerobic digestion plant at the compound of the Addis Ababa University (Katharina Panse 2013)

3.1.2. Canteen and Kitchen Waste Feed

As the feed of the biogas plant, the kitchen waste and food left over are used. The waste is collected by the kitchen people and then transported to the reception tank (Figure 3.2) of the AD plant once a day. The wastewater from the kitchen is delivered through water pipes to a wastewater tank underneath the kitchen. There, it can be stored until needed. A water hose that is connected to the bottom of the tank is used to transport the wastewater from the wastewater tank to the AD plant. Depending on the actual amount of solid waste, wastewater is added to the reception tank. The waste is mixed and crushed inside the tank through a granulation pump. A feeding pump delivers the substrate automatically from the reception tank into the fermenter.

Figure3.2: Transport of kitchen waste to reception tank (Katharina Panse 2013)

Through anaerobic digestion, the substrate is turned into biogas inside of the fermenter. The produced biogas flows from the gas dome through a gas meter and a gas analyser to the gas storage. The gas storage is almost pressure-less and located side by side to the fermenter. The volume of the gas holder is 8.8m^3. The gas holder is made of a gas dense coated polyester textile foil especially constructed for biogas.

After reaching a certain filling level in the gas holder, the generated biogas is ready for usage. By activating the gas request through a switch inside the kitchen, a gas blower starts working and a magnetic valve opens the gas pipe so that the biogas is delivered directly to the kitchen.

If the maximum filling level of the gas storage is reached, surplus biogas will be burned by an emergency flair. Therefore the gas blower starts again, but this time a second magnetic valve is activated. The second magnetic valve opens the gas pipe to the emergency flare. The flare is automatically started by ignition plugs and burns the unused biogas innocuously.

In order to make the kitchen independent of Eucalyptus wood a biogas cooker had been installed within the dish washing room. The cooker is designed to heat up water for dish washing and the gas system had been construed to cover the daily demand for hot water of the kitchen.

3.1.3. Absorption column

A small absorption column is built from long tubular glass. The column has a dimension of height 250 cm and diameter 20 cm. The absorption column is filled with glass packing to create a larger contact surface between the gas and water. The gas flow rate is measured by using Ritter drum type gas meter (type, TG10). The apparatus measures both the gas pressure and flow rate. The biogas is pumped from biogas storage into the absorption column by air compressor.

3.1.4. Digital Gas Analyzer, SR2-BIO, Gas analyzer

Directly connected to the silka gel gas holder to measure the gases (CH_4 in Vol %, CO_2 in Vol %, O_2 in Vol% and H_2S in ppm)

3.1.5. Silka gel container vessel

The silka gel container contains a silka gel and the purpose is to store the product gas and absorb the moisture before it goes to the gas analyzer.

3.2 Methods

3.2.1 Batch counter-current biogas upgrading experiment

The water washing or biogas upgrading experiment was conducted at the Addis Ababa University, Addis Ababa Institute of Technology student’s dormitory compound after preparing a pilot scale counter-current water wash upgrading facility. The water wash unit consisted of a vertical absorption column glass, 2.5m length with an inner diameter of 20cm. Both sides of the absorption column have two holes which were used for water inlet and gas outlet in the upper side while the bottom holes used vice-versa.

First the raw gas was channeled into the gas storage from the fermenter. After the raw gas is filled into the gas storage, the raw gas was analyzed prior to starting the experiment. The raw gas and absorbent (in this case, water) were then channeled into the absorption column in which the gas flow rate, water flow rate and the water head are maintained.

The absorbent used in the process was tap water (pH 7) at an average inlet temperature of $11.5^{O}C$. Water was sprayed from the top of the column and the raw gas was compressed using an air compressor from the bottom of the column. After the upgrading has taken place in the absorption column, the product gas which contains concentrated methane, other unrefined gases and moisture were channeled into the silka gel vessel. In this vessel the high moisture added to the biogas compounds due to the absorbent water will be absorbed by the silka gel before it goes to the gas analyzer which could be damaged due to the high moisture in the gas.

Upgrading efficiency was studied using different water head (80,110 and 140 cm) and water (8, 16 and 25 l/min) and gas (1, 1.5 and 2 l/min) flows in trials. The gas analyzer was set on for each batch experiment and the product gas composition was continuously recorded and the data was taken when the reading become stable.

3.2.2 Estimation method for Methane Enrichment

Methane enrichment efficiency from each upgrading experiment results were calculated using the following equation:

$$Enrichment\ CH4 = 100 * \frac{Cp-Cr}{Cr} \qquad (3.1)$$

Where,

Cp: is methane content in product gas, and

Cr: is methane content in raw gas

Carbon dioxide and hydrogen sulphide removal efficiency has been calculated as well.

3.2.3 Study variables

The absorption column was arranged based on the objective set to identify the influence(s) of major factors. These factors studied in this study were:

- Water flow rate: 8, 16 and 25 l/min
- Gas flow rate: 1, 1.5 and 2 l/min
- Water head: 80, 110 and 140 cm

3.3. Statistical Analysis

In order to evaluate the effect of the three main factors (biogas flow rate, water flow rate and water head) independently from each other and their interactions effect, ANOVA was run by means of Design-Expert® 7.1 software. Among different designs presented in this software factorial design (general factorial) was selected because this approach enabled the inclusion and analysis of multiple factors at multiple levels. Responses under consideration were: methane enrichment or methane upgrading efficiency, CO_2 and H_2S trapping or removal efficiency.

A confidence interval of 95% (P=0.05) was used, so all statistical analysis were compared and treated as being significantly different, if the *P*-value was less than 0.05. The entire collected ANOVA results for each response are presented in Appendix C.

4. RESULT AND DISCUSSION

In this section, we discussed on the findings obtained from the biogas upgrading experiment. The experimental set up is outlined in Chapter 3. The experiment involves absorption of carbon dioxide and hydrogen sulfide from biogas produced by biogas pilot plant situated in AAiT student dormitory campus. The experiment uses water as an absorbent and tested with glass packing materials. Different experimental or operational parameters were investigated to see the effects of the parameters on removal of carbon dioxide and hydrogen sulfide from the biogas in methane enrichment. In the section below, the experiment executed and their results are presented, analyzed and discussed.

4.1 Raw biogas analysis

All the measurements of the raw biogas used in this study were carried out in the Addis Ababa Institute of Technology student dormitory using SR2-BIO, digital gas analyzer. The raw data obtained here were also used to calculate methane enrichment efficiency using the upgraded results.

As prescribed in Chapter 3, water scrubbing absorption pilot scale plant was constructed to execute the experiment. The column is packed with glass packing. The column was used to generate twenty seven runs with three replicates.

When each run was conducted, the raw biogas was analyzed at the start of the experiment, in the middle of the experiment and just before experiment is finished for every batch experiment. Such analysis was done due to the significant variation in composition of the gas so that the average result would be taken. The analysis equipment measures the composition of the three main components (CH_4, CO_2 and H_2S) of raw biogas and oxygen and the results are presented in Table 4.1. The upgraded gas composition was analyzed just before the end of the experiment.

As it can be seen from Table 4.1, the methane content was very low due to the higher amount of carbon dioxide produced. This high amount of carbon dioxide decreases the utilization of methane as an energy source. The amount of hydrogen sulfide is also significant. In order to improve the quality of the biogas energy content, carbon dioxide and hydrogen sulfide need to be removed.

Table 4.1 Feed Biogas composition as a function of water and gas flow rates and water head

WF (l/min)	GF (l/min)	WH(cm)	CH_4 (vol. %)	CO_2 (vol. %)	H_2S (vol. %)*
8	1	80	45.33	50.67	0.0249
16	1	80	56.33	40.67	0.020367
25	1	80	53.33	43.67	0.020867
8	1.5	80	55	42	0.0162
16	1.5	80	52.67	43	0.023667
25	1.5	80	54.33	40.67	0.0318
8	2	80	54	42	0.041267
16	2	80	55	40.33	0.032333
25	2	80	52.33	45	0.0351
8	1	110	56.67	40.67	324.67
16	1	110	53.67	42.33	0.024533
25	1	110	56	40.33	0.021867
8	1.5	110	56.33	39.67	0.0278
16	1.5	110	55	40.67	0.024067
25	1.5	110	53.67	42.67	0.023833
8	2	110	54	41	0.015833
16	2	110	53.67	41.67	0.028867
25	2	110	53.33	42.33	0.029833
8	1	140	55.33	41	0.036267
16	1	140	55	41.67	0.0389
25	1	140	56.67	40.33	0.0437
8	1.5	140	53	41.67	0.0410
16	1.5	140	53.67	42.33	0.024833
25	1.5	140	54.67	41.33	0.0218
8	2	140	56	41.67	0.0197
16	2	140	53	44.33	0.023533
25	2	140	56	41	0.024967

***** The H_2S composition was first read in ppm and here converted to vol. %. The sum is not 100% because beside the main components, biogas also contains other sulphide compounds, siloxanes, aromatic and halogenated compounds, oxygen and nitrogen

4.2 Biogas upgrading experiment

4.2.1 Absorption performance of column with glass packing material

The methane composition upgrading and carbon dioxide reduction due to the scrubbing of carbon dioxide and hydrogen sulfide were measured for different operation parameters: water flow rate (WF), gas flow rate (GF) and water head (WH). The absorption column filled with glass packing was used. The result obtained is present in Table 4.2.

Table 4.2: Experimental results for biogas upgrading as function of water flow rates, gas flow rates and water heads

WF (l/min.)	GF (l/min.)	WH (cm)	CH_4 (vol. %)	CO_2 (vol. %)	H_2S (vol. %)
8	1	80	61.33	33	0.0035
16	1	80	64.33	31	0.002133
25	1	80	66.33	31	0.003133
8	1.5	80	61.33	35.33	0.0032
16	1.5	80	63	33.67	0.003267
25	1.5	80	66.33	30.33	0.0034
8	2	80	60.66	35.33	0.0027
16	2	80	60	33.33	0.002433
25	2	80	62.33	32.33	0.002633
8	1	110	65.67	31.33	0.002633
16	1	110	67	26.33	0.002667
25	1	110	77	22.67	0.0022
8	1.5	110	62.67	31	0.002033
16	1.5	110	67.67	29	0.0030
25	1.5	110	78	18.67	0.003222
8	2	110	62.33	35.33	0.0027
16	2	110	63.67	30.33	0.0023
25	2	110	67.33	27	0.002567
8	1	140	66.67	28	0.0020
16	1	140	70	25	0.0021
25	1	140	77.67	17	0.0024
8	1.5	140	64	31.33	0.0022
16	1.5	140	72.33	21	0.001833
25	1.5	140	79.67	16.33	0.001933
8	2	140	64	30.33	0.002067
16	2	140	72	21.67	0.002167
25	2	140	77	17.67	0.0019

***** The H_2S composition was first read in ppm and here converted to vol. %. The sum is not 100% because beside the main components, biogas also contains other sulphide compounds, siloxanes, aromatic and halogenated compounds, oxygen and nitrogen. The amounts of trace compounds are low compared to the main components.

4.2.1.1 Effects of water heads, water and gas flows

Suppose A stands for water flow rate (WF), B stands for raw biogas flow rate (GF), and C stands for water head (WH). The methane content of the upgraded gas (scrubbed biogas) was significantly affected by A, B and C and the interaction AC (interaction of water flow rate and water head).

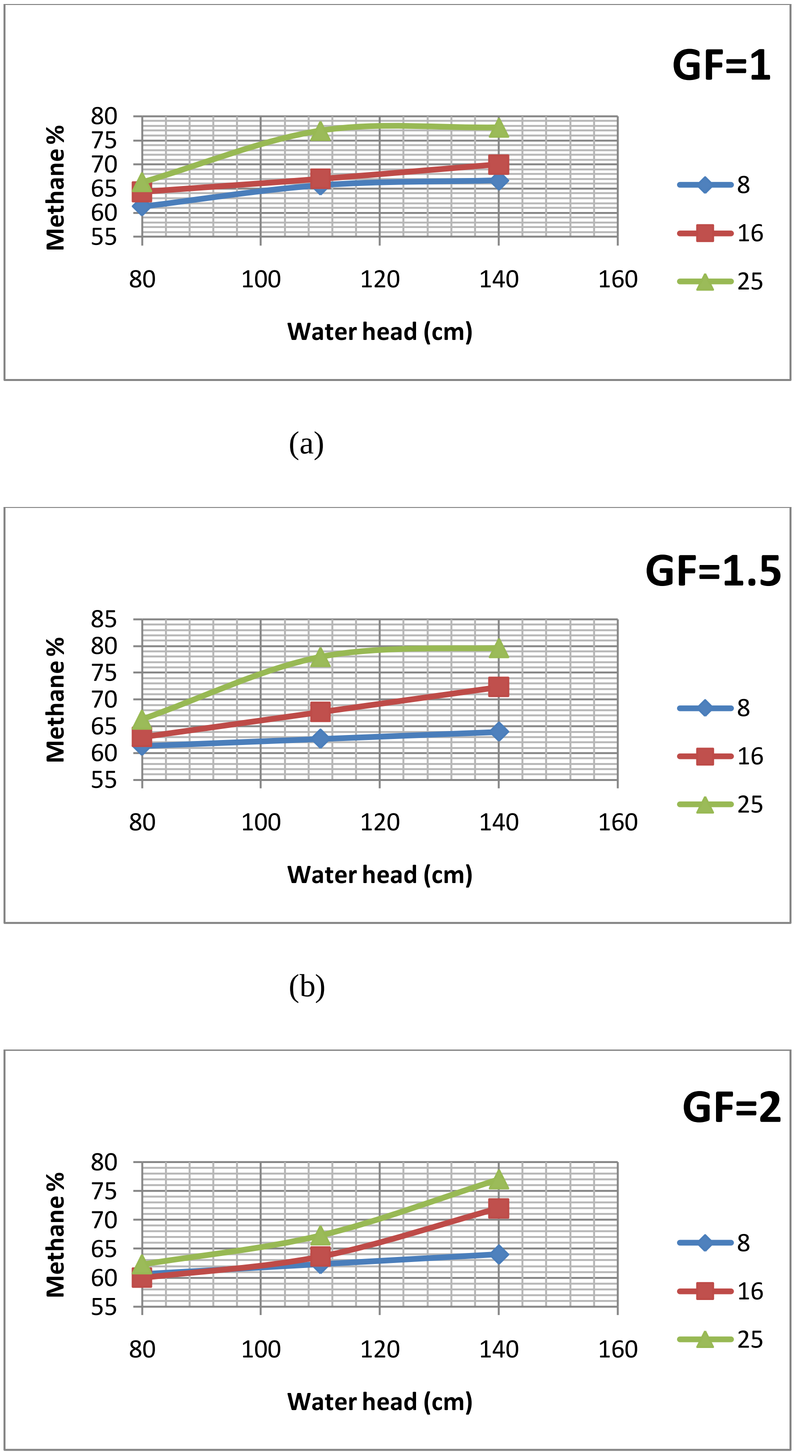
The effect of varying water head on methane content of the product gas is presented in Figure 4.1. It can be seen from Figure 4.1 (a, b and c) that, when the water head increased, so did the concentration of methane in the product gas (upgraded biogas) which is scrubbed with water at a flow rate of 8, 16 and 25 l/min. For instance: Figure 4.1/a shows, a significant increase in the methane content when water flow changes. The increase in water head increased the methane concentration. For instance, at GF= 1 l/min and WF = 25 l/min, increase of WH from 80 cm to 140 cm increases methane content from about 66 % to about 78 %. In Figure 4.1/b, the methane content reaches almost 80 % at the same head and water flow (140cm and 25 l/min respectively) when the gas flow was increased by 0.5, from 1 to 1.5 l/min. The methane concentration tends to decrease though it is very small. For instance at WF =8 l/min and WH= 140 cm, concentration of methane decreases from 66 % to 62 % when GF increased from 1 to 2 l/min. This can be seen in Figure 4.1/c.

In consistent to these results Rasi (2009) reported that the methane content of the product gas was close to or above 90 % at over 20 bar pressure with 10 l/min water flow and at 30 bar pressure with 5 l/min water flow. This report clearly showed that higher pressure with lower water flow or relatively lower pressure with higher water flow was selected due to its high upgrading efficiency. The results in Figure 4.1 show that there is increment in product gas methane content in all cases, but the product gas contained over 75 % methane at high water head and water flow. According to Rasi (2009), the higher pressure was selected to increase the product gas concentration as it favors the carbon dioxide and hydrogen sulphide solubility in water , in this research a higher water head is selected for the same reason as the pressure since the gas develops high pressure inside the absorber as it goes up to higher head. Therefore the reason for the increment of methane in the product gas as the water head increases is the result of high water head that helped to develop high upgrading pressure which on the other hand makes the solubility of carbon dioxide and hydrogen sulphide higher keeping the absorbent used in the process (tap water) at a temperature of 12-14.5$^{O}_{C}$.

Another reason for higher concentration of methane in the product gas is likely due to the packing material. This has helped to increase the area of contact between the liquid and the gas phases in a way favoring mass transfer.

Interaction Effect of Water Head and Water flow rate:

It is clearly seen in Figure 4.1 that for all the three biogas flows which is to be scrubbed (1, 1.5 and 2 l/min), the highest upgraded methane was obtained when the raw biogas is treated with the highest water head and water flow (140cm and 25 l/min respectively) while the lowest methane was obtained when the scrubbing is done at the lowest water head (80 cm) and lowest water flow (8 l/min). The results showed that an interaction of higher water head and water flow rate gives higher methane content in the upgraded product gas. The reason here also is that the highest head and highest water flow are able to develop higher upgrading pressure which is efficient for absorption.



(a)

(b)

(c)

Figure 4.1: Effect of water head and gas flows on methane composition of product gas after washing the raw biogas at 8 l/min (a), 16 l/min (b) and 25 l/min (c) water flows

Purification Performance of Column vs WF/GF

Figure 4.2 illustrates that the methane composition generally increases as liquid-to-gas ratio increases. Solubility is a very important factor affecting the amount of a pollutant that can be absorbed. The more soluble the pollutant gas, the less the amount of liquid required. Liquid-to-gas ratios for gas absorption are often higher, (3 to 6 liters per actual m³) (Hobler, 1966). A minimum volume of liquid is required to create sufficient collection (scrubbing) targets.

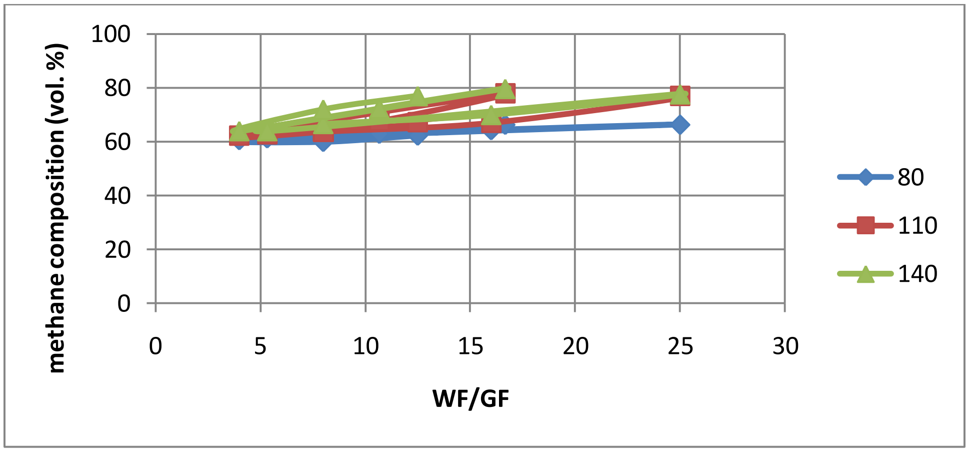
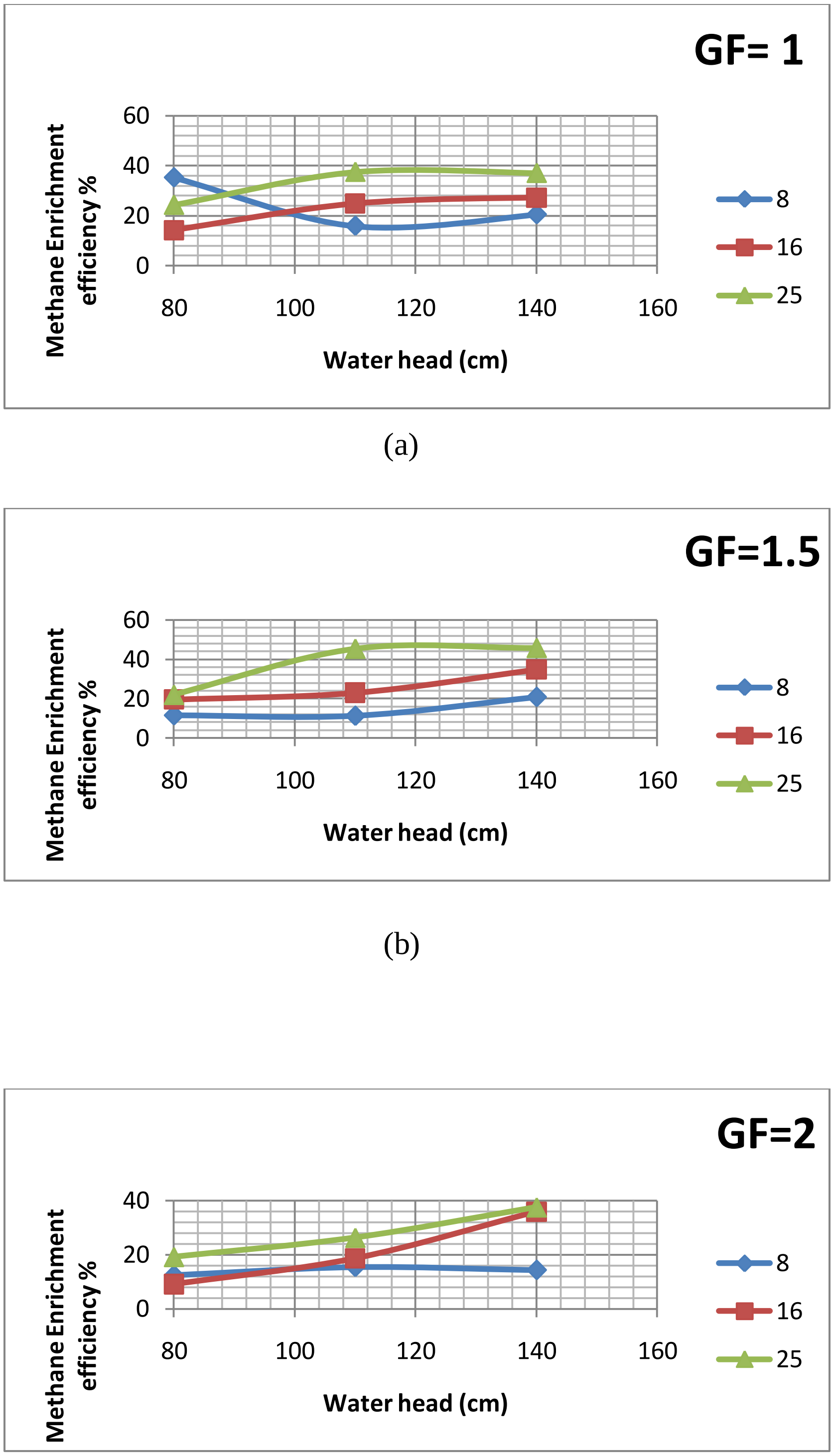


Figure 4.2: Effects of water head and liquid-to-gas ratios on purification performance of column

From this study, it can be learnt that methane enrichment efficiency could be calculated using the methane composition in the product gas. The results as presented in Figure 4.3 shows that the methane enrichment efficiency increases for all the three biogas flows (1, 1.5 and 2 l/min). Where the highest enrichment efficiency was obtained when the absorption is conducted at higher water head and water flow (140 cm and 25 l/min respectively) and when the gas flow is (1.5 l/min). As it has been presented in Figure 4.1/b above, the highest methane content in product gas is obtained when the gas is treated with (140cm and 25l/min water head and water flow respectively), implying methane enrichment is reached highest when the upgrading methane is higher.



(a)

(b)

(c)

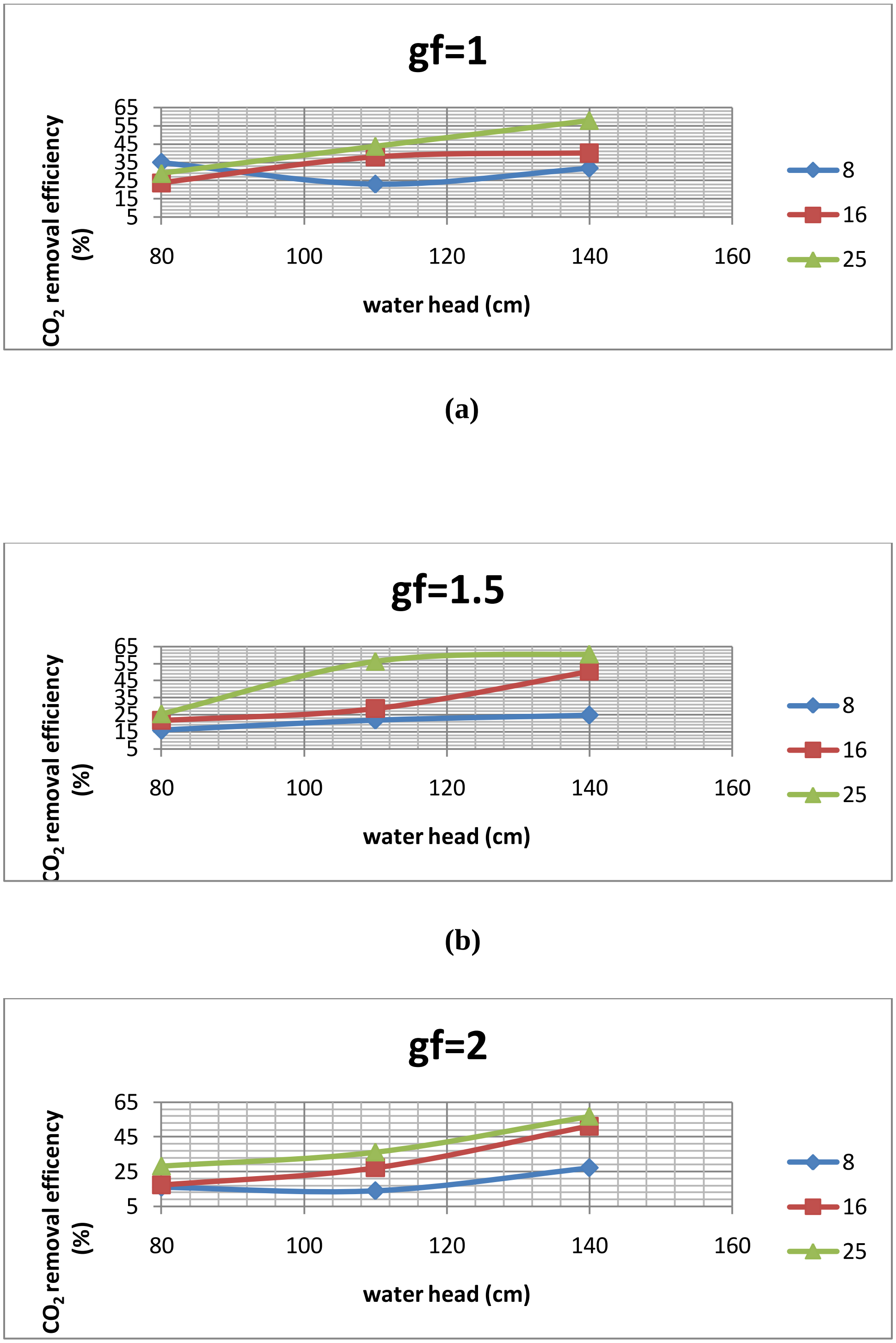
Figure 4.3: The effects of water head, water flow rate of 8, 16 and 25 l/min on methane enrichment efficiencies treated under a gas flow of 1, 1.5 and 2 l/min

Carbon Dioxide Removal Efficiency

On the other hand removal of carbon dioxide in the raw gas was studied using the same parameters and the main factors A (water flow rate), B (raw biogas flow rate), C (water head) and the interaction AC (interaction of water flow rate and water head) have significant ($p<0.05$) effect on CO_2 removal (Appendix C.2).

As it is represented graphically in Figure 4.4 below (a, b and c), carbon dioxide removal efficiency increases as the water head increases. The highest value of CO_2 removal was observed in biogas treated with 140 cm water head, while the lowest values were recorded at lowest water head (80 cm).

Similar to this study, substantial increase in carbon dioxide removal at higher upgrading pressure was also observed by Rasi (2009). In all the gas/water ratios studied, the product gas contained less than 4.5% carbon dioxide which implies above 90% carbon dioxide removal was achieved. While high proportion of carbon dioxide (over 60% removal efficiency), was observed in this study resulting in CH_4 enriched biogas.



(a)

(b)

(c)

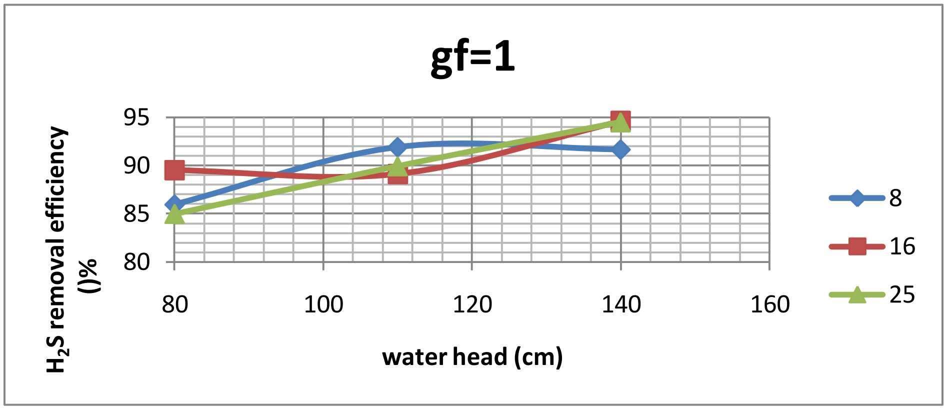
Figure 4.4: Effect of water head, scrubbing water flow, raw biogas flow and interaction between water head and water flow rate on CO_2 removal efficiency

Hydrogen sulphide removal efficiency

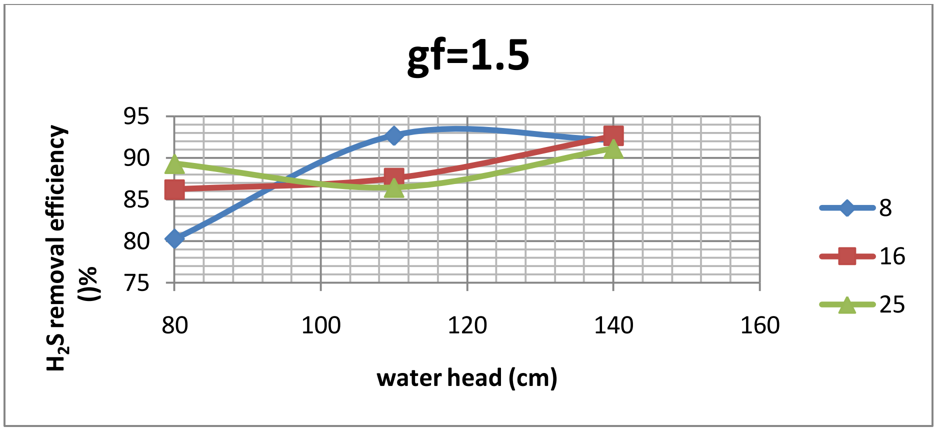
The statistical analysis showed that water head level has significantly ($p<0.05$) affect the hydrogen sulphide solubility rate in water. The effect of water head level increment on H_2S removal efficiency is presented in Figure 4.5 below.

The hydrogen sulphide content of the raw biogas ranged from 162 to 437 ppm was removed from the product gas at much higher removal efficiency in all conditions studied (Figure 4.5). The gas removal efficiency was increased as the water head level increases and the highest value of removal efficiency was recorded in all conditions which is above 90% (Figure 4.5).

Unlike carbon dioxide, highest hydrogen sulphide removal for all the conditions is observed. The reason for this is the highest solubility rate of the H_2S gas compared to CO_2. The observation is consistent with a study conducted by (Lide 1996). According to his finding an increase in H_2S removal could be resulted from water washing which is an effective upgrading process for biogases produced from different sources as carbon dioxide ($8.21 * 10^{-4}$, 288.15K) and hydrogen sulphide ($2.335 * 10^{-3}$, 288.15K) are more soluble in water than methane ($3.122 * 10^{-5}$, 288.15 K). Among these three gases H_2S is by far more soluble.



(a)



(b)

Figure 4.5: Effect of water head, water flow rate and raw biogas flow rate on H_2S removal efficiency

4.3. The difference in energy value between the raw biogas that is being used in the pilot plant and its increment in energy due to the purification

In this study the analysis was made to find and compare the energy value of the upgraded biogas with the raw biogas. Unitrove software was used to calculate the calorific value of both the scrubbed and raw biogas. Where the average value of the triplicates data for the twenty seven run is used for the scrubbed biogas and for the raw biogas the same manner is followed as the gas composition was measured three times for a batch experiment.

The software presents the result in many measuring units like (KJ/mol, MJ/Kg, MJ/m^3, KWh/ Kg), the megajoule per cubic meter which shows the useful energy in megajoules released from one cubic meter of the biogas is selected to see the difference in energy value of the raw and upgraded biogas. It's clearly seen in Table 4.3 below that the pilot plant upgrading has brought about a significant increment in energy value as the scrubbing leads to a 45.6% increase in calorific value.

Table 4.3: Calorific value comparison for raw and upgraded biogas

Raw CH_4 (vol %)	Upgraded CH_4 (vol %)	Calorific value of raw biogas (Mj/m^3)	Calorific value of upgraded biogas (Mj/m^3)	Calorific value increment (%)
45.33	61.33	17.15	23.2	35.28
56.33	64.33	21.31	24.33	14.17
53.33	66.33	20.17	25.08	24.34
55	61.33	20.80	23.19	11.50
52.67	63	19.92	23.82	19.58
54.33	66.33	20.55	25.08	22.05
54	60.66	20.43	22.94	12.29
55	60	20.80	22.69	9.09
52.33	62.33	19.80	23.57	19.04
56.67	65.67	21.43	24.83	15.87
53.67	67	20.30	25.33	24.79
56	77	21.18	29.11	37.44
56.33	62.67	21.31	23.70	11.22
55	67.67	20.80	25.59	23.03
53.67	78	20.30	29.48	45.22
54	62.33	20.43	23.57	15.37
53.67	63.67	20.30	24.08	18.62
53.33	67.33	20.17	25.46	26.23
55.33	66.67	20.93	25.21	20.45
55	70	20.80	26.46	27.21
56.67	77.67	21.43	29.36	37.00
53	64	20.05	24.20	20.70
53.67	72.33	20.30	27.34	34.68
54.67	79.67	20.68	30.11	45.60
56	64	21.18	24.20	14.26
53	72	20.05	27.22	35.76
56	77	21.18	29.11	37.44

4.4 Biogas Water Heating Trial

A test was conducted to see how long it would take to boil 2 liters of water using a biogas before upgrading (raw biogas) and after it has been upgraded to the highest methane content in the product gas. Since water heating is a very energy intensive process, it would help to see how hot it would get for the given time and be able to visualize the physical meaning of upgrading biogas.

The 2 liter water reached 75 degrees in 19 minutes when the raw biogas (56 vol. %) was applied as energy source for heating and the same volume of water reached 75 degrees in 8 minutes faster when the highest methane content (80 vol. %) in upgraded biogas was used for heating.

This study confirm from the water heating trial that there is a significant time difference in heating water with a raw biogas (56 vol. %) and upgraded biogas (80 vol. %).

5. CONCLUSIONS AND RECOMMENDATIONS

5.1. Conclusions

This study was conducted to investigate the effect of upgrading biogas (a pilot plant biogas produced at AAIT student dormitory) using water scrubbing technology to increase the biogas potential in terms of its energy value for better energy utilization.

The results of this study indicate that the biogas upgrading using water scrubbing is influenced by water head and water and gas flow rates. Increasing water head, gas and water flow increases the methane content in the product gas. This on the other hand implies that the amount of carbon dioxide and hydrogen sulphide removal increases as the three parameters increases.

The biogas composition analysis conducted at various water heads (80, 110 and 140 cm) has indicated that the methane concentration showed a significant increment in the upgraded biogas. As the water head increases the two water soluble gases (carbon dioxide and hydrogen sulphide) has been led to a continuous decrease in their volume in product gas. The highest value of biogas upgrading efficiency for methane was attained at 140 cm head with the highest CO_2 and H_2S removal efficiency. Highest upgrading efficiency or highest removal efficiency of impurities was found as the water head increases due to the higher solubility pressure acquirement. Therefore, having long absorption column is essential in order to increase methane enrichment capacity.

Even though, biogas upgrading studies were not conducted in Ethiopia so far whether water scrubbing is cheaper and simpler or not but it can be taken from different countries experience that this technology is better for those who got plenty of water resources.

5.2. Recommendations

Efficient use of energy resources is an important aspect of energy management. Upgrading raw biogas to high methane content is one form in which biogas energy resource can be efficiently used, and in a sustainable manner. As noticed from the present pilot plant study, the methane content of the pilot plant gas can be upgraded to 80% methane by a counter-current water absorption process. During upgrading the carbon dioxide content reduced below 17 % and hydrogen sulphide was removed with high efficiency (90 %).

Due to its potential and feasibility for cooking, electricity generation and different other purposes the biogas needs to be purified for easy storage into cylinders and prolong the efficiency of various equipments used for biogas application purposes. Depending on the biomethane standards applied in different countries, upgraded biogas could be used in several biogas applications, including vehicle fuel.

The findings of this pilot plant upgrading system adds to the evidence that biogas may used as a renewable energy source and a substitute for fossil fuels. However, biogas enrichment will be much improved if further research work is conducted considering the following issues:

- In this research the absorption column used was not designed according to the desired concentration output of CH_4, CO_2 and H_2S. So that in order to get desired output concentration of the gases, calculations should be made from the mass transfer equations and the minimum flow rate of water from the given raw biogas flow rate and also the minimum contact area required to get the mass transfer should be determined.
- In order to select the highest pressure for upgrading due to its high upgrading efficiency, different runs at deferent pressures should be conducted. To do this biogas compressor with which pressure could be varied need to be in place.
- It might be pertinent if biogas is produced in larger volumes to study carbon dioxide mixture with other contaminants after purification process in order to suggest on feasibility of carbon dioxide extraction to supply or sell for use for different purposes since carbon dioxide content is about 40 % of total biogas production on site.

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Appendix

Appendix A: Calorific value results

A.1. Calorific value results for the feed raw biogas

Table A.1. Calorific value results for the feed raw biogas

CH_4 (vol. %)	**CO_2 (vol. %)**	**H_2S (vol. %)**	**KJ/mol**	**MJ/kg**	**MJ/m3**	**KWh/kg**
45.33	50.67	0.0249	404.14	13.13	17.15	3.65
56.33	40.67	0.020367	502.22	18.13	21.31	5.04
53.33	43.67	0.020867	475.47	16.66	20.17	4.63
55	42	0.0162	490.36	17.47	20.8	4.85
52.67	43	0.023667	469.58	16.35	19.92	4.54
54.33	40.67	0.0318	484.38	17.14	20.55	4.76
54	42	0.041267	481.44	16.98	20.43	4.72
55	40.33	0.032333	490.36	17.47	20.8	4.85
52.33	45	0.0351	466.55	16.19	19.8	4.5
56.67	40.67	324.67	505.25	18.31	21.43	5.08
53.67	42.33	0.024533	478.5	16.83	20.3	4.67
56	40.33	0.021867	499.27	17.97	21.18	4.99
56.33	39.67	0.0278	502.22	18.13	21.31	5.04
55	40.67	0.024067	490.36	17.47	20.8	4.85
53.67	42.67	0.023833	478.5	16.83	20.3	4.67
54	41	0.015833	481.44	16.98	20.43	4.72
53.67	41.67	0.028867	478.5	16.83	20.3	4.67
53.33	42.33	0.029833	475.47	16.66	20.17	4.63
55.33	41	0.036267	493.3	17.63	20.93	4.9
55	41.67	0.0389	490.36	17.47	20.8	4.85
56.67	40.33	0.0437	505.25	18.31	21.43	5.08
53	41.67	0.0410	472.53	16.51	20.05	4.59
53.67	42.33	0.024833	478.5	16.83	20.3	4.67
54.67	41.33	0.0218	487.42	17.31	20.68	4.81
56	41.67	0.0197	499.27	17.97	21.18	4.99
53	44.33	0.023533	472.53	16.51	20.05	4.59
56	41	0.024967	499.27	17.97	21.18	4.99

A.2. Calorific value results for the upgraded biogas

Table A.2. Calorific value results for the upgraded biogas

CH_4 (vol. %)	CO_2 (vol. %)	H_2S (vol. %)	KJ/mol	MJ/kg	MJ/m3	KWh/kg
61.33	33	0.0035	546.79	20.54	23.2	5.71
64.33	31	0.002133	573.54	22.37	24.33	6.21
66.33	31	0.003133	591.37	23.75	25.08	6.60
61.33	35.33	0.0032	546.79	20.79	23.19	5.78
63	33.67	0.003267	561.68	21.75	23.82	6.04
66.33	30.33	0.0034	591.37	23.75	25.08	6.60
60.66	35.33	0.0027	540.82	20.42	22.94	5.67
60	33.33	0.002433	534.94	20.06	22.69	5.57
62.33	32.33	0.002633	555.71	21.36	23.57	5.93
65.67	31.33	0.002633	585.49	23.34	24.83	6.48
67	26.33	0.002667	597.35	24.17	25.33	6.71
77	22.67	0.0022	686.5	31.33	29.11	8.70
62.67	31	0.002033	558.74	21.55	23.70	5.99
67.67	29	0.0030	603.32	24.60	25.59	6.83
78	18.67	0.003222	695.42	32.14	29.48	8.93
62.33	35.33	0.0027	555.71	21.36	23.57	5.93
63.67	30.33	0.0023	567.66	22.14	24.08	6.15
67.33	27	0.002567	600.29	24.38	25.46	6.77
66.67	28	0.0020	594.40	23.96	25.21	6.66
70	25	0.0021	624.09	26.14	26.46	7.26
77.67	17	0.0024	692.47	31.87	29.36	8.85

64	31.33	0.0022	570.60	22.33	24.20	6.20
72.33	21	0.001833	644.87	27.77	27.34	7.71
79.67	16.33	0.001933	710.31	33.56	30.11	9.32
64	30.33	0.002067	570.60	22.33	24.20	6.20
72	21.67	0.002167	641.92	27.53	27.22	7.65
77	17.67	0.0019	686.50	31.33	29.11	8.70

Appendix B: result for WF to GF ratio

Table B.1. Result for WF to GF ratio

WF/GF	Water head (cm)	CH4 (vol. %)
8	80	61.33
16	80	64.33
25	80	66.33
5.33	80	61.33
10.67	80	63
16.67	80	66.33
4	80	60.66
8	80	60
12.5	80	62.33
8	110	65.67
16	110	67
25	110	77
5.33	110	62.67
10.67	110	67.67
16.67	110	78
4	110	62.33
8	110	63.67
12.5	110	67.33
8	140	66.67
16	140	70
25	140	77.67
5.33	140	64
10.67	140	72.33
16.67	140	79.67
4	140	64
8	140	72
12.5	140	77

Appendix C: Design Expert 7.0.0 Software Output

C.1. ANOVA result for CH_4 upgrading analysis

Table C.1. ANOVA result for the response CH_4 upgrading

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	Remark
Model	839.14	7	119.88	26.70	< 0.0001	significant
A-water flow rate	385.96	1	385.96	85.96	< 0.0001	
B-gas flow rate	40.21	1	40.21	8.96	0.00075	
C-water head	342.33	1	342.33	76.25	<0.0001	
AB	5.13	1	5.13	1.14	0.2985	
AC	64.96	1	64.96	14.47	0.0012	
BC	5.08	1	5.08	1.13	0.3008	
ABC	3.26	1	3.26	0.73	0.4049	
Residual	85.31	19	4.49			
Cor Total	924.45	26				

C.2. ANOVA result for CO_2 removal analysis

Table C.2. ANOVA result for the response for CO_2 removal

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	Remark
Model	847.33	7	121.05	34.94	< 0.0001	significant
A-water flow rate	337.21	1	337.21	97.34	< 0.0001	
B-gas flow rate	17.84	1	17.84	5.15	0.0351	
C-water head	428.31	1	428.31	123.64	<0.0001	
AB	0.39	1	0.39	0.11	0.7422	
AC	68.00	1	68.00	19.63	0.0003	
BC	3.34	1	3.34	0.96	0.3384	
ABC	0.025	1	0.025	0.733	0.9327	
Residual	65.82	19	3.46			
Cor Total	913.15	26				

C.3. ANOVA result for H2S removal analysis

Table C.3. ANOVA result for the response H_2S removal

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	Remark
Model	360.62	7	51.52	3.25	0.0193	significant
A-water flow rate	0.97	1	0.97	0.061	0.8076	
B-gas flow rate	9.37	1	9.37	0.59	0.4517	
C-water head	337.34	1	337.34	21.26	0.0002	
AB	0.0004719	1	0.0004719	.000002975	0.9957	
AC	0.18	1	0.18	0.012	0.9152	
BC	3.09	1	3.09	0.19	0.6639	
ABC	8.83	1	8.83	0.56	0.4648	
Residual	301.42	19	15.86			
Cor Total	662.04	26				